Spectroscopy of atomic rubidium at 500 bar buffer gas pressure: approaching the thermal equilibrium of dressed atom-light states

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We have recorded fluorescence spectra of the atomic rubidium D-lines in the presence of several hundreds of bars buffer gas pressure. With additional saturation broadening a spectral linewidth comparable to the thermal energy of the atoms in the heated gas cell is achieved. An intensity-dependent blue asymmetry of the spectra is observed, which becomes increasingly pronounced when extrapolating to infinitely high light intensity. We interpret our results as evidence for the dressed (coupled atom-light) states to approach thermal equilibrium.

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In usual optical spectroscopy experiments, light sources along with spontaneous decay drive the investigated sample far from thermal equilibrium [1]. This is most evident in room temperature atomic physics experiments, where the observed spectral linewidths in energy units are many orders of magnitude below the thermal energy k_BT ($\simeq$ 1/40 eV for T = 300 K). Thus, apart from a comparatively small and symmetric broadening arising from the known Doppler effect, the excitation profile is largely independent of the statistical distribution function. In cold atom experiments, spectral linewidth and thermal energy can be comparable. However, the lack of a sufficiently fast thermalization process has so far here prevented thermal equilibrium of coupled atom-light states to be a useful concept.

We here report on experiments, in which up to 500 bar of argon and helium respectively buffer gas pressure induce a linewidth of a few nanometers for the rubidium D-lines. At high optical power of the exciting continuous-wave laser source, the fluorescence spectra are broadened by additional power broadening to values exceeding the thermal energy k_BT in the heated gas cell. In this regime, we observe a strong blue asymmetry of the spectra. The spectral asymmetry increases further when extrapolating our data towards infinitely high excitation intensity. We interpret our results as evidence for the coupled atom-light states ("dressed states") to approach thermal equilibrium, with the thermalization being due to frequent rubidium-buffer gas collisions. Thermal equilibrium is e.g. a prerequisite for possible BEC-like phase transitions of coupled atom-light quasiparticles (polaritons) [2].

Our experiment benefits of collisional aided excitation, which allows to saturate the atomic transition at large detuning with a continuous-wave laser source. Pressure broadening of atomic spectral lines is a long investigated topic [3, 4, 5, 6, 7, 8, 9, 10]. For large laser detunings ($\gg 1/\tau_{coll}$, where τ_{coll} denotes the collisonal duration), the impact limit ceases to be fulfilled, and atomic lines often become asymmetric. A theoretical description of the absorption probability requires knowledge of the molecular potential curves of the collisional partners [11], and can be expressed e.g. in terms of modulated dipole approaches [12]. The effect of collisional redistribution has shown the influence of state changing collisions [13]. In an interesting experiment, deviations from the Einstein coefficients for absorption and stimulated emission have been observed for far off-resonant excitation in a collisionally broadened system[14]. Dressed state approaches have proven to be an elegant way to treat collisional processes in the presence of laser radiation [15].

For a theoretical description of the transition to thermal equilibrium of dressed states, we give a simple model based on thermodynamic arguments. Consider a two-level system with ground state $|g\rangle$ and excited state $|e\rangle$ coupled to a laser field of frequency ω. When tuned into resonance, the laser field connects the states $|g, n+1\rangle$ and $|e, n\rangle$ respectively, where the first quantum number denotes the internal atomic state and the second one the photon number. To simplify the analysis, let us restrict ourselves to the case of large detunings δ = ω - ω_{atom}, with $|\delta| \gg \Omega_{Rabi}$, so that in the absence of collisions both the state mixing and the AC Stark shift can be neglected. The energy splitting between dressed states is thus simply δ, as shown in Fig. 1a. On the other hand, when a buffer gas atom approaches the energy levels are shifted, and efficient excitation and a transfer between dressed state energy levels is provided when the laser field transiently becomes resonant during the collision, yielding an example of collisional aided excitation. Let $c_{g\rightarrow e}(\delta)$ and $c_{e\rightarrow g}(\delta)$ denote the rate constants for collisional transfer between the dressed state levels, which for $|\delta| \gg \Omega_{Rabi}$ can be shown to be proportional to the laser intensity [15]. For a finite spontaneous decay rate Γ, also a coupling between states $|e, n\rangle$ and $|g, n\rangle$ of different dressed state manifolds occurs.

We are interested in the properties of such a two level system subject to laser radiation and coupled to a thermal bath. If we assume our system to be optically thin, we can trace over the photonic quantum numbers, and obtain the following Boltzmann-like rate equations

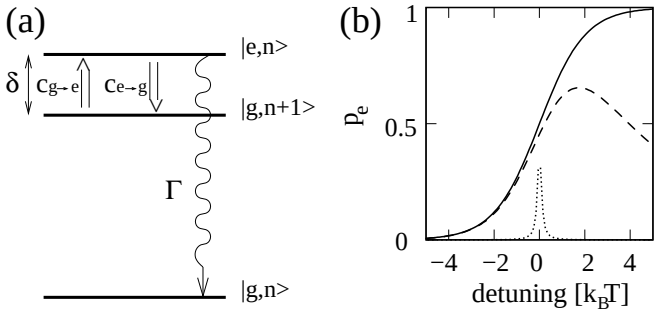

FIG. 1: (a) Scheme of levels and rates for a two-level dressed state system approaching thermal equilibrium in the far detuned limit. (b) Upper state population as a function of laser frequency, where a Lorentzian lineshape was taken for the detuning dependent excitation rate $c(\delta)$: For $c(k_BT) \ll \Gamma$, i.e. small linewidth, which yields a lineshape resembling that of $c(\delta)$ (dotted line), $c(k_BT) \simeq \Gamma$ (dashed line) and $c(\delta) \gg \Gamma$, i.e. thermal equilibrium of dressed states (solid line).

$$\dot{f}_e = c_{g\to e}(\delta) f_g - c_{e\to g}(\delta) f_e - \Gamma f_e \tag{1a}$$

$$\dot{f}_g = c_{e\to g}(\delta) f_e - c_{g\to e}(\delta) f_g + \Gamma f_e, \tag{1b}$$

where f_e and f_g are the corresponding populations of states $|e\rangle$ and $|g\rangle$. To derive a relation between the rates $c_{g\to e}$ and $c_{e\to g}$, let us consider the equilibrium solution ($\dot{f}_e = \dot{f}_g = 0$) in the absence of spontaneous decay (i.e. $\Gamma \to 0$). In this limit, we expect that usual Boltzmann statistics can be applied to this two-level system, which yields $f_e = e^{\delta/k_BT}/(1+e^{\delta/k_BT}) = 1/(1+e^{-\delta/k_BT})$, which resembles the Fermi-Dirac distribution function due to only two available states. From Eqs. 1 we obtain $f_e = 1 - f_g = 1/(1 + c_{e\to g}/c_{g\to e})$ so that the following relation of the rate constants must apply: $c_{e\to g}/c_{g\to e} = e^{-\delta/k_BT}$, as also shown in [14]. Note that the sign of the laser detuning in this formula depends on the definition of δ. Physically, the increased rate towards a population of the energetically lower dressed state is due to the larger phase space that becomes available to the reservoir with the associated energy gain of $|\delta|$. The result is also obtained when considering the variation of the available density of states in Fermi's golden rule.

Spontaneous decay drives the dressed state system out of thermal equilibrium. The full Eq. 1 with $\dot{f}_e = \dot{f}_g = 0$ yields the following stationary solution in our model:

$$f_e = \frac{c(\delta)/\Gamma}{1 + \frac{c(\delta)}{\Gamma} \cdot (1 + e^{-\delta/kT})} \tag{2}$$

The condition for this formula to reduce to the usual Fermi-Dirac distribution of a two-level system at large transition linewidth and laser intensity is that $c(\delta) \gg \Gamma$ is fulfilled at all relevant detunings δ. On the other hand, for $k_BT \gg |\delta|$ we obtain $f_e \simeq (c(\delta)/\Gamma)/(1 + 2c(\delta)/\Gamma)$, which corresponds to the usual saturation dependence of a two-level system [15]. Fig. 1b shows a comparison of line profiles in the different regimes, where a Lorentzian excitation profile was assumed for the detuning dependent excitation rate $c(\delta)$. The dotted line gives the spectrum in the limit of a linewidth of $c(\delta)$ considerably below k_BT, resulting in a lineshape resembling that of $c(\delta)$, while the solid line gives the case of a linewidth considerably above the thermal energy (solid line). Also, an example for the intermediate regime of comparable linewidth and thermal energy is given (dashed line). A theoretical prediction of the detuning dependent excitation rate $c(\delta)$ in the here investigated dense buffer gas regime is a highly nontrivial task, especially since we expect multiple particle collisions to play an important role. On the other hand, if full thermal equilibium can be achieved for all used laser detunings, the predicted lineshape reduces to a Fermi-Dirac profile independently of the form of $c(\delta)$.

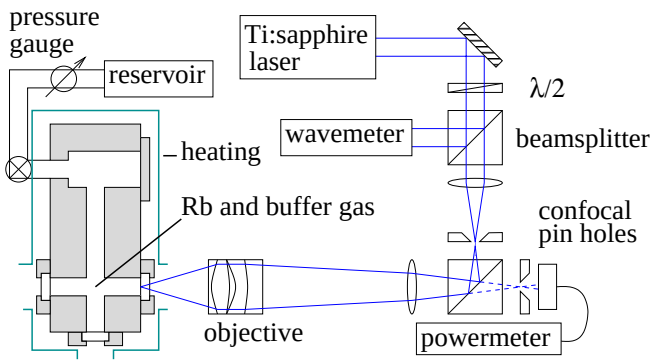

FIG. 2: Scheme of the experimental setup.

A scheme of our experimental setup used to investigate rubidium atoms at high buffer gas pressures and intense laser radiation is shown in Fig. 2. A rubidium filled stainless steel oven with volume of a few cm^3 and wall thickness of roughly a cm is used as a pressure cell. Optical radiation is coupled into the chamber through sapphire windows. Buffer gas is filled into the chamber through an attached valve with pressures up to 230 bar (at room temperature). Due to technical reasons, the pressure is measured outside the chamber. During operation, the cell is heated to 260 °C, which results in a vapour pressure limited rubidium number density of $\simeq 1.0 \cdot 10^{16}$/cm^3 [16]. In the sealed chamber, the buffer gas pressure then increases up to approximately 400 bar for helium and 500 bar for argon.

Tunable laser radiation for excitation of the atoms is provided by a Ti:sapphire ring laser. Laser frequency scanning over the broad atomic absorption profile is done manually within the operation range of the used laser mirror set of 350-395 THz. The laser frequency is measured with a wavemeter. With a Fraunhofer achromat, the laser radiation is focused into the rubidium chamber to a beam radius of ~ 3 μm, which at a maximum optical power of 300 mW results in an intensity of $1.1{\cdot}10^8$ mW/cm^2. The beam focal plane within the cell is placed near the sapphire window. To selectively record the

atomic fluorescence only from regions of high laser intensity, we furthermore spatially filter both the incident optical beam and the outgoing fluorescence with pinholes in a confocal geometry. The recorded fluorescence power values exhibited short-term variations that became increasingly severe for a focal spot very close to the cell window, which attribute to thermal fluctuations. For our experimental spectra, the mean fluorescence power value averaged over 30 seconds is taken for every data point.

In initial experiments, we have measured the lifetime of the rubidium 5P state at high argon buffer gas pressures. For this measurement, an acoustooptic modulator (not shown in Fig. 2) was included in the beam path, which allowed for a rapid shutting of the exciting optical radiation for up to $\sim$ 30 mW beam power. The subsequent decay of the atomic fluorescence was monitored with a photomultiplier tube. At low buffer gas pressures ($\lesssim$ 10 bar) we observed fluorescence decay times up to 1 μs, which we attribute to repopulation from higher states due to energy pooling [17]. More direct evidence for energy pooling is obtained from the clearly visible blue fluorescence at those pressure values. At higher buffer gas pressures the fluorescence signal decayed with a time constant of 40 ns. If we account for the time constants of our acoustooptic modulator and the photomultiplier detection, this value is in good agreement with the lifetime value of about 27 ns obtained in earlier measurements [18]. We conclude that energy pooling to higher state excitation is suppressed at sufficient buffer gas pressure. Further, the lifetime of the $5P_{1/2}$ and $5P_{3/2}$ states are, within our measurement accuracy of 5 ns, unchanged to argon pressures of 200 bar, which is the maximum achieved pressure value in these early data sets. At a collisional rate of 10^{11}/s, the atoms experience more than 10^3 collisions within a natural lifetime. This confirms the remarkable elasticity of excited atoms with rare gas collisions [19], and is a prerequisite for the thermalization of dressed states in our experimental scheme. Note that the excited state lifetime here is some four orders of magnitude above the $\sim$ps lifetimes observed in semiconductor exciton systems [2].

Typical fluorescence spectra recorded under high pressure buffer gas conditions for variable light intensities are shown in Fig. 3a (argon buffer gas) and Fig. 3b (helium buffer gas). While the observed lineshapes somewhat differ for both used buffer gases, indicating the 'transiently chemical' nature of optical collisions in a regime beyond the impact limit, let us draw our attention to some general features for both spectra. At moderate optical power ($P \simeq 25$ mW) the linewidths are clearly below the thermal energy k_BT ($\simeq$ 11 THz at $T = 530$ K). At higher power levels the linewidths increase to values around k_BT. Noticeably, the red side of the D1-line tends to saturate to a comparatively low fluorescence level, while at the blue side of the D2-line much higher fluorescence is observed, though in the far wings even at

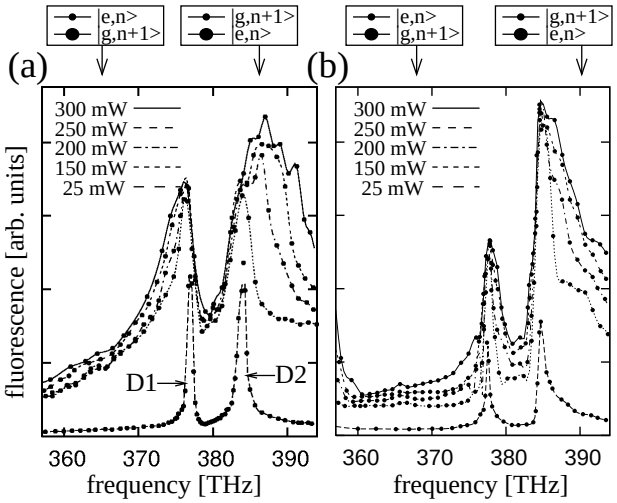

FIG. 3: Fluorescence spectrum of the rubidium D-lines at 500 bar argon (a) and 400 bar helium (b) buffer gas pressure for different optical beam powers. The small drawings on the top indicate the population of the dressed states on the red and blue side of the electronic transition respectively.

maximum power levels (300 mW) the transition is less saturated. We attribute this to the dressed state system approaching thermal equilibrium for high power levels. The resonant Rabi coupling at full optical power is of order 100 GHz, so that for detunings of order k_BT the mixing of ground and excited states is still small. For the sake of simplicity, let us restrict the discussion to the red side of the D1-line and the blue side of the D2-line, as in these limits the influence of the upper state fine structure splitting is smallest. While on the red side of a two-level system the state $|g, n+1\rangle$ is energetically below $|e, n\rangle$, leading to an enhanced ground state population in thermal equilibrium, on the blue side of the transition the dressed excited state component is lowest and expected to be favored in equilibrium, as indicated in the boxes in the top of Fig. 3. At low optical power levels, the upper state spontaneous decay drives the coupled atom light system away from equilibrium. For a more detailed analysis, Fig. 4a shows the observed fluorescence as a function of the optical power for three different laser frequencies for the data with helium buffer gas. These saturation curves have been fitted with the theoretical prediction of Eq. (2), where a constant prefactor and the ratio of $c(\delta)$ and the laser intensity were left as free fit parameters. This fitting procedure assumes that the detuning dependent excitation rate $c(\delta)$ is linearly dependent on the laser intensity, an assumption in agreement with present theories of optical collisions, whose validity in the yet largely unexplored high pressure buffer gas regime with large multiparticle collisional rate however remains to be tested [21]. Near a line center (crosses), the curve saturates already at relatively low power level to an intermediate fluorescence level. For significant de-

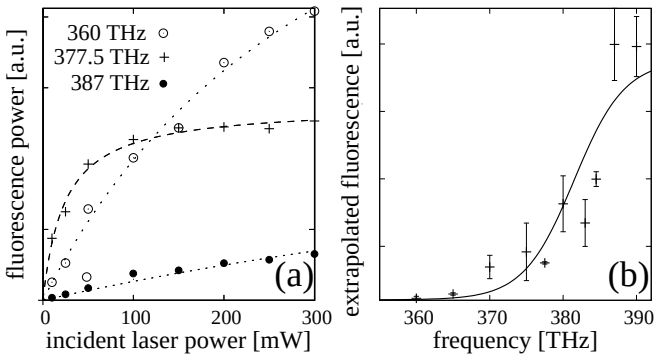

FIG. 4: (a) fluorescence of rubidium atoms at 400 bar helium buffer gas as a function of optical power for different laser frequencies. The experimental data has been fitted with the expected saturation dependence of Eq. (2). (b) Expected atomic fluorescence at infinite laser power, obtained from extrapolation curves as in (a), versus laser frequency. The shown error bars were obtained from the extrapolation fits. The data has been fitted with a Fermi-Dirac distribution, assuming thermal equilibrium of the two-level dressed state system.

tuning the optical power at which saturation is achieved is higher, as visible both for blue (dots) and red (circles) detunings. Interestingly, the saturation level of fluorescence is clearly different in both cases, with the blue (red) detuning leading to largest (smallest) values.

Fig. 4b shows the value of fluorescence extrapolated to infinite beam power, as taken from such curves, as a function of laser frequency. This is to provide an estimate for the upper state population at thermal equilibrium. The data has been fitted with a Fermi-Dirac distribution $f_e(\delta) = 1/(1 + e^{-\delta/k_B T})$ expected for the upper state population of the two level dressed state system, where the detuning δ was measured relatively to the center of the D-lines. The agreement between theory and experiment is quite reasonable, which supports the assumption of the dressed states to approach thermal equilibrium at high laser power. An issue remaining to be clarified is that a best fit is obtained with a temperature $T = 162(58)$K, which is significantly below the cell temperature. For a comparable fit with the argon buffer gas spectral data, a temperature of 270(91)K was obtained. At present, the origin of this disagreement of temperatures is not resolved. A more detailed rate equation model of partial thermal equilibrium should include the rubidium fine structure and spatially inhomogeneous saturation. On the other hand, an interesting question is whether laser cooling (heating) processes of the thermal gas here occur on the red (blue) side of the transition due to the energy loss (gain) during collisional aided excitation, as was proposed in [20]. If the spontaneously emitted photons are energy shifted by an amount comparable to $k_B T$ respectively to the incident photon wavelength, cooling may be possible in such pressure broadened systems, comparable to results obtained on the optical cooling of solids [22].

To conclude, we have recorded spectra of rubidium atoms in a collisionally broadened regime interpolating between usual atomic physics gas phase and solid/liquid phase conditions. With additional saturation broadening, the spectral linewidth approaches the thermal energy. The saturation dependence of the line profile is interpreted as evidence for the onset of thermal equilibrium of dressed atom-light states.

For the future, it would be interesting to study possible novel laser cooling mechanisms of high pressure atomic gases based on collisionally aided excitation. A quite different perspective could include a possible BEC-like phase transition to a condensed atom-light polariton phase. For our present apparatus, this would require an increase of the optical intensity and an optically thick atomic ensemble, as could be realized with a cavity- or optical waveguide based system.

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